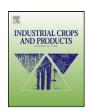
ELSEVIER

Contents lists available at SciVerse ScienceDirect

## **Industrial Crops and Products**

journal homepage: www.elsevier.com/locate/indcrop



# Modification of yield and composition of essential oils by distillation time

Jeffery B. Cannon<sup>a</sup>, Charles L. Cantrell<sup>a</sup>, Tess Astatkie<sup>b</sup>, Valtcho D. Zheljazkov<sup>c</sup>,\*

- a Natural Products Utilization Research Unit, Agricultural Research Service, U.S. Department of Agriculture, P.O. Box 8048, University, MS 38677, United States
- <sup>b</sup> Department of Engineering, Nova Scotia Agricultural College, P.O. Box 550, Truro, NS B2N 5E3, Canada
- c Mississippi State University, North Mississippi Research and Extension Center, 5421 Highway 145 South, Verona, MS 38879, United States

#### ARTICLE INFO

#### Article history: Received 8 October 2011 Received in revised form 28 February 2012 Accepted 11 April 2012

Keywords: Essential oil Distillation Lemongrass Peppermint Palmarosa

#### ABSTRACT

Altering the distillation times of economically important essential oils such as peppermint ( $Men-tha \times piperita$  L.), lemongrass (Cymbopogon flexuosus Steud.), and palmarosa (Cymbopogon martinii Roxb.) oils may allow producers to increase the production, engineer the composition, and decrease the energy required for distillation. Experiments were conducted to model essential oil yield and oil composition of peppermint, lemongrass, and palmarosa oils as a function of the length of the steam distillation time (DT). Maximum essential oil yields of peppermint, lemongrass, and palmarosa were achieved at a DT of 20 min; further increases in DT did not increase oil yields. In lemongrass and palmarosa experiments, DTs of 240 min led to 25–40% reductions in oil yield compared to yields at 20–160 min. This study demonstrated that DT can be used as a tool for obtaining essential oils with specific targeted composition from peppermint, lemongrass, and palmarosa. Secondly, the study found that the optimum length of the DT for maximum essential oil yields of peppermint, lemongrass, and palmarosa was much shorter than the time usually used by researchers and processors. Shorter DT may save producers and processors energy costs and other resources. This study also suggests that comparison of data on oil composition must take into consideration the length of the DT.

© 2012 Elsevier B.V. All rights reserved.

## 1. Introduction

Peppermint, lemongrass, and palmarosa essential oils are used in many economic applications. Peppermint oil is used widely in aromatic applications such as toothpaste, mouthwash, and chewing gum, but it also has important culinary and pharmaceutical applications (Lawrence, 2007). Oil of lemongrass has wide uses in the culinary, pharmaceutical, and cosmetics industries; it is also considered an eco-friendly insect-repellant (Ganjewala and Luthra, 2010; Weiss, 1997). Along with uses in traditional medicine, palmarosa oil is an effective anti-helminthic (Kumaran et al., 2003) and insect repellant for grains and legumes (Kumar et al., 2007). Understanding the effects of distillation time (DT) on essential oil (EO) yield and composition may allow producers of these economically important oils to increase the production and engineer the composition of the oils while decreasing the energy required for distillation.

E-mail addresses: vjeliazk@uwyo.edu, valtcho.pubs@gmail.com (V.D. Zheljazkov).

There is no previous comprehensive study on the effect of DT on peppermint, lemongrass, and palmarosa oils; although there are some analogous studies on oil yield and composition. Rohloff et al. (2005) found that peppermint oil yield, but not overall composition, varied with pre-distillation treatments (ground wilting and drying temperatures), but this study did not vary DTs. Katsiotis et al. (1985) reported that varying DTs (1–4 h) and distillation rates (0.5, 2.0, 3.5, and 5.0 ml/min) had no significant effect on oil yield and quality; using intact peppermint leaves during distillation was the only important factor in determining peppermint oil yield and quality. The method of distillation used also can affect yield and quality of the oils (Scheffer, 1993).

Various factors influencing peppermint EO yield have been studied, whereas lemongrass and palmarosa oils are somewhat understudied in this regard. One study of lemongrass using steam distillation found that EO yield increased with increasing DT (following first-order kinetics) and when plant material was packed loosely into the distiller, however, no results of the effects of DT on oil composition were presented (Koul et al., 2004). Another study of steam distillation documented changes in lemongrass oil composition with field nitrogen and sulfur application, but this study did not investigate various distillation treatments (Zheljazkov et al., 2011). Our literature search showed no previous reports on the effects of DT for palmarosa oil; however, citronella oil (derived from a congeneric species, *Cymbopogon* 

<sup>\*</sup> Corresponding author. Current address: University of Wyoming, Sheridan Research and Extension Center, 663 Wyarno Road, Sheridan WY 82801, United States. Tel.: +1 307 737 2415; fax: +1 307 737 2413.

winterianus Jowitt) was examined in one study (Cassel et al., 2006). These authors reported a maximum oil yield (0.942%, wt/wt) with steam distillation of fresh plant material for 4h (Cassel et al., 2006).

Another issue that needed to be addressed is the fact that various researchers used different DTs for extracting oils from peppermint, lemongrass, and palmarosa. It is not known if reports with different DTs would be comparable, and, if so, within what time limits.

The objective of this study was to use steam distillation and subsequent gas chromatography/mass spectrometry (GC/MS) analysis of the resulting essential oil to elucidate how commercial yield and quality of peppermint, lemongrass, and palmarosa oils might be affected by varying the DT of the plant material. Knowledge of optimum DT may help producers of these oils conserve energy and resources while improving oil quality and yield. Furthermore, it may be possible to use DT as a tool for controlling and influencing EO composition. In addition, such study would provide the basis for comparison of published studies using different DTs.

#### 2. Materials and methods

#### 2.1. Growing conditions

Peppermint, lemongrass, and palmarosa plants for this study were produced in field experiments conducted in 2010 at the North Mississippi Research and Extension Center at Verona, MS (34°43′22″N and -88°43′22″W).

#### 2.1.1. Peppermint

In this study, we used the peppermint variety traditionally grown in the United States, 'Black Mitcham'. 'Black Mitcham' comprises around 90% of peppermint plantations in the US. All commercial peppermint varieties are propagated vegetatively because peppermint is a natural hybrid (Zheljazkov et al., 2010b). Transplanting of peppermint in the field was done with certified virus-free transplants purchased from Summit Plant Laboratories, Ft. Collins. CO. in two rows on a raised bed, with 30 cm of space between the plants. Growing environment and harvesting conditions for peppermint were as described previously (Zheljazkov et al., 2010b). Plants were grown in a randomized complete block design for four years (2007-2010); samples for this study were taken in 2010. Harvesting was done at flowering, and the plants were dried at temperature not exceeding 40 °C to avoid essential oil losses, and to reflect the common practice in US peppermint industry where plants are harvested and left to dry for 3 or 4 days before distillation.

## 2.1.2. Lemongrass and palmarosa

Lemongrass and palmarosa transplants were started from certified seed purchased from Richters (Ontario, Canada). Transplant production and growth conditions were as described previously (Zheljazkov et al., 2011). Briefly, transplants were produced in controlled environment greenhouse, in plastic trays with growth medium (Metromix 300, Scotts Co., Marysville, OH). When the plants were approximately 12 cm tall, they were transplanted into the field. Lemongrass and palmarosa were planted in raised beds covered with black plastic, and with irrigation drip tape under the plastic. Plants were spaced 60 cm apart, in a randomized complete block design with 4 replicates. Plants were harvested in August 2010, when both species reached technical maturity. Subsamples from lemongrass and palmarosa were taken by harvesting the plants at 12–15 cm above the soil surface. Harvest was done in the morning, and subsamples of 500 g of fresh material were immediately subjected to distillation; the lag time between harvest and beginning of the distillation was not more than 45 min.

#### 2.2. Steam distillation and distillation times (DTs)

Dried peppermint samples (200 g dried material), and fresh lemongrass and palmarosa samples (500 g), all in 3 replicates, were steam distilled in 2-L steam distillation units (Hearthmagic, Rancho Santa Fe, CA), as described previously (Zheljazkov et al., 2010a, 2010b, 2011). The apparatus we used resembles that used by commercial distillers. Essentially, the apparatus consists of a 2-L pear-shaped flask filled with water on a hotplate. Above this flask is positioned the plant material in a 2-L reservoir (bioflask) open on top and bottom. The stillhead is attached to the top of this bioflask and directs the steam to the condenser which allows for the codistilled steam and oil to collect and separate simultaneously in a specially designed funnel, similar to Florentine vessel (separator) in commercial installations. The results from these 2-L systems are relevant to larger commercial type distillation equipment. This statement is supported by previous research (with another team) in which we compared distillation yield and oil profile of the steam distillation research units used in this study to semiindustrial distillation units for several crops including peppermint and lemongrass, but did not find differences in oil yield or oil composition between smaller 2-L system and semi-industrial systems. The DTs for the peppermint samples were 1.25 min, 2.5 min, 5 min, 10 min, 20 min, 40 min, 80 min, and 160 min, whereas the DTs for lemongrass and palmarosa samples were 1.25 min, 2.5 min, 5 min, 10 min, 20 min, 40 min, 80 min, 160 min, and 240 min. The distillation times were selected to include the range of distillation times reported in the literature and on our preliminary experiments. These distillation times were measured from the beginning of the distillation, at the moment when the first drop of essential oil was

The resulting essential oils from each distillation were separated from water in the Florentine part of the apparatus, and the last inseparable drops were separated by freezing the water. The amount of essential oil was measured on an analytical scale, and the essential oil content was calculated as grams of oil per weight of dry peppermint or fresh lemongrass or palmarosa tissue. The essential oil samples were kept at  $-5\,^{\circ}\text{C}$  until GC/MS analyses were performed.

# 2.3. Gas chromatography–mass spectroscopy (GC/MS) analysis of essential oil

Chemical standards and peppermint, lemongrass, or palmarosa essential oils were analyzed by GC/MS on a Varian CP-3800 GC coupled to a Varian Saturn 2000 MS/MS (Palo Alto, CA). The GC was equipped with a CP Sil 8CB fused silica capillary column (30 m  $\times$  0.25 mm, with film thickness of 0.25  $\mu m$ ) operated using the following conditions: injector temperature, 240 °C, column temperature, 60–120 °C at 3 °C/min followed by 120–240 °C at 20 °C/min then held at 240 °C for 5 min; carrier gas, He; injection volume, 1  $\mu L$  (splitless). The MS mass ranged from 40 to 650 m/z, filament delay of 3 min, target TIC of 20,000, a prescan ionization time of 100  $\mu s$ , an ion trap temperature of 150 °C, manifold temperature of 60 °C, and a transfer line temperature of 170 °C.

Geraniol, *n*-dodecane, caryophyllene oxide, citral (*cis-trans* mixture), (-)-*trans*-caryophyllene, and *n*-tridecane were purchased from Sigma–Aldrich (St. Louis, MO). Eucalyptol, menthofuran, (-)-menthol, (-)-menthone, (-)-(1R)-menthyl acetate and geranyl acetate were purchased from Fluka (Switzerland). Retention times for each analyte and internal standard were as follows in minutes in parentheses: geraniol (18.375), *n*-dodecane (16.000), caryophyllene oxide (24.436), *Z*-citral (17.690), *E*-citral (19.016), (-)-*trans*-caryophyllene (22.674), *n*-tridecane (20.263), eucalyptol (9.340), menthofuran (14.454), (-)-menthol (14.871),

(-)-menthone (14.015), (-)-(1R)-menthyl acetate (19.925) and geranyl acetate (22.070).

With five to six concentration points, an external standard least squares regression (Bates and Watts, 2007) was used to confirm linearity in analyte response factor. All analytes were used to formulate separate calibration curves. The total ion chromatograms of each of the oils from the distillation experiments were compared to the standard injections. The target peaks were confirmed by retention time and mass spectral data. Confirmed integrated peaks were then used to determine the percentage of each chemical constituent in the essential oil based on an internal standard using the calculation method by Magee and Herd (1999). The internal standard used for peppermint and palmarosa oils was *n*-dodecane, and the internal standard used for lemongrass was *n*-tridecane.

A solution of CHCl $_3$  containing the appropriate internal standard was prepared at a concentration of 0.1 mg/mL. Using a micropipet, 12–17  $\mu$ L of essential oil from each sample was transferred into a 10-mL volumetric flask. Samples were brought to volume with CHCl $_3$  containing internal standard. This or a similar method was used to prepare 1.0 mg/mL concentrations of palmarosa and peppermint essential oils and 1.5 mg mL $^{-1}$  concentrations of lemongrass essential oil containing the appropriate internal standard. A 1 mL portion of each oil sample was placed by glass pipet into a GC vial for analysis as described above.

## 2.4. Statistical analysis

The effect of DT on essential oil content, and the concentration and yield of constituents was determined using a one-way analysis of variance (ANOVA). For each response, the validity of model assumptions was verified by examining the residuals as described in Montgomery (2009). For the responses that were significantly affected by DT (at the 5% level of significance), multiple means comparison was completed using Duncan's multiple range test at the 5% level of significance. The analysis was completed using SAS (SAS Institute Inc., 2008).

The most appropriate model that describes the relationship between DT and essential oil content as well as the concentrations of key constituents was identified to be either a Third-order polynomial model (Eq. (1)), the Asymptotic (concave) model (Eq. (2)), or the Power (convex) model (Eq. (3)).

$$Y = \beta_0 + \beta_1 X + \beta_2 X^2 + \beta_3 X^3 + \varepsilon \tag{1}$$

$$Y = \theta_1 - \theta_2 e^{(-\theta_3 X)} + \varepsilon \tag{2}$$

$$Y = \theta_1 X^{\theta_2} + \varepsilon \tag{3}$$

where Y is the response variable and X is DT, the independent variable. The error term  $(\varepsilon)$  is assumed to be distributed normally and independently with constant variance. Since the Asymptotic (concave) and the Power (convex) models are nonlinear, the parameters were estimated iteratively (Bates and Watts, 2007) using the NLIN procedure of SAS (SAS Institute Inc., 2008).

#### 3. Results

Due to the complexity of this study and the large number of components present in essential oils, it was necessary that we focused on the major and most economically important constituents present in the peppermint, lemongrass, or palmarosa essential oils. In peppermint essential oil, we performed absolute quantification of the constituents menthol, menthofuran, (-)-menthone, eucalyptol, (-)-(1R)-menthyl acetate, and (-)-trans-caryophyllene (Fig. 1). For lemongrass essential oil we chose geranial (E-citral), neral (E-citral), caryophyllene oxide, and E-caryophyllene (Fig. 2)

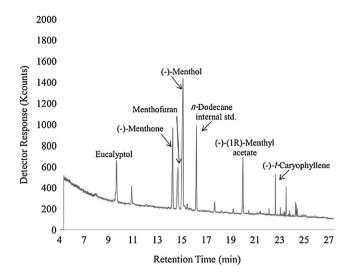
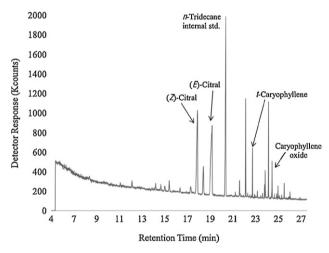
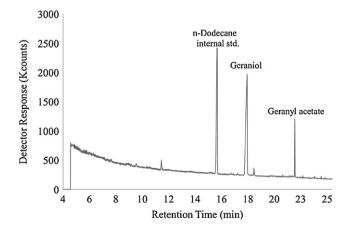


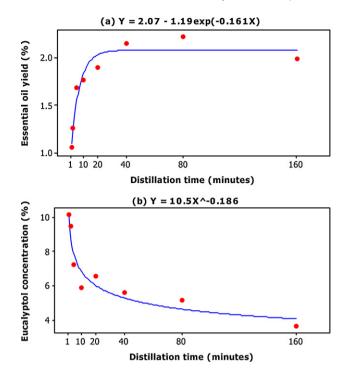
Fig. 1. Representative GCMS total ion chromatogram of *Mentha* × *piperata*, peppermint essential oil.



**Fig. 2.** Representative GCMS total ion chromatogram of *Cymbopogon flexuosus*, lemongrass essential oil.



**Fig. 3.** Representative GCMS total ion chromatogram of *Cymbopogon martinii*, palmarosa essential oil.



**Fig. 4.** Peppermint – plot of DT vs. essential oil content and concentration of eucalyptol along with the fitted (a) Asymptotic (concave) for essential oil content, and (b) Power (convex) for eucalyptol concentration non-linear regression models. The fitted models are given as sub-titles.

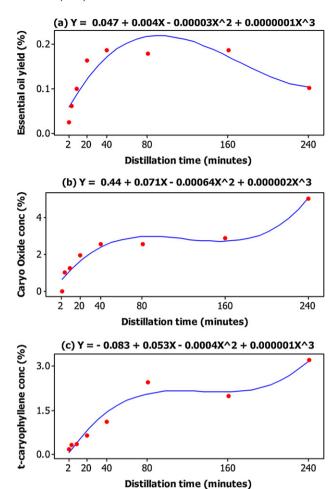
whereas for palmarosa essential oil we examined the two constituents geraniol and geranyl acetate (Fig. 3).

#### 3.1. Effects of distillation time (DT) on peppermint oil

Peppermint essential oil yield increased rapidly initially and then leveled off, with a yield starting at 1.06% in the 1.25 min DT and maximizing near 2% in the 20-160 min DT (Table 1). The change in essential oil yield as a function of DT was very well modeled by the Asymptotic (concave) non-linear regression model (Fig. 4). The concentration of two of the peppermint major constituents, menthol and menthone, was not affected by the DT. Another major oil constituent, menthofuran concentration was lowest in the oil from the 1.25 DT and highest in the 40 or 160 min DT (Table 1). Menthyl acetate, one of the minor components of peppermint oil, was not affected by DT (Table 1). Eucalyptol was the only component that showed a significant and steady decrease in concentration with increasing DT (from 10% at 1.25 min DT to 3.7% at 160 min DT (Table 1; Fig. 4). This decrease was adequately modeled by the Power (convex) non-linear regression model (Fig. 4). Unlike eucalyptol, t-caryophyllene gradually increased with DT, ranging from 1.8% at 1.25 min DT to 3.7% at 160 min DT. Generally, the yield of individual constituents (a function of the concentration and the oil yield) increased until 20 or 40 min DT. However, the yield of eucalyptol remained the same, indicating that eucalyptol generally was eluted off during the first few minutes of the distillation process.

#### 3.2. Effects of distillation time (DT) on lemongrass oil

Lemongrass essential oil yield increased and reached a maximum at 20 min DT (Table 2; Fig. 5). We observed no significant differences in oil yield in the 20–160 min distillation treatments. The oil yield at 240 DT actually decreased by about 40% relative to the yield at 20–160 min DT, most probably due to evaporation (Table 2; Fig. 5). The concentration of the major components



**Fig. 5.** Lemon grass – plot of DT vs. essential oil yield and concentrations of *t*-caryophyllene and caryophyllene oxide (caryo oxide) together with fitted third order (cubic) polynomial regression model. The fitted models are given as sub-titles.

of lemongrass essential oil, neral and geranial, were lowest at the shortest DT, generally higher at 5–40 min DT compared to the shortest DT, and generally decreased again beyond 40 min DT (Table 2). Caryophyllene oxide concentrations generally increased with increasing DT and ranged from being below the limit of quantification at 1.25 min to the maximum of 5% by 240 min (Table 2; Fig. 5). However, the yield of caryophyllene oxide maximized at 40 min DT; there was no further increase with longer DT. *t*-caryophyllene generally increased with increasing DT and reached maximum at 240 min DT (Table 2; Fig. 5). However, *t*-caryophyllene yield maximized much earlier, at 80 min DT and did not change after that (Table 2). The relationship between DT and essential oil yield as well as the concentrations of caryophyllene oxide and *t*-caryophyllene was adequately modeled by a cubic polynomial regression model (Fig. 5).

## 3.3. Effects of distillation time (DT) on palmarosa oil

Palmarosa essential oil yield followed similar trends as lemongrass and peppermint with DT. Oil yield was 0.009% in the 1.25 min DT and reached maximum near 0.19% in the 20 min DT (Table 3; Fig. 6). Palmarosa oil yield was not significantly different at DTs of 20 min, 40 min, 80 min, or 160 min, but decreased by about 25% at 240 min apparently due to evaporation. The mean concentration of geraniol, the major palmarosa oil constituent, ranged between 65.2 and 71.4% and was not affected significantly by the DT. However, geranyl acetate concentration significantly increased

**Table 1**Mean peppermint essential oil yield, and the concentrations of eucalyptol, menthofuran, menthol, menthone, menthyl acetate, and *t*-caryophyllene obtained from the 8 distillation times (DT).

DT (min)	EO content (%)	Concentration of oil constituents as % of the total oil							
		Eucalyptol	Menthofuran	Menthol	Menthone	Menthyl acetate	t-Caryophyllene		
1.25	1.06d	10.1a	10.9c	17.4a	14.9a	7.1a	1.8c		
2.50	1.26d	9.5ab	16.0abc	18.2a	16.3a	7.0a	2.6bc		
5	1.68c	7.3abc	13.8bc	18.7a	14.3a	7.3a	2.8abc		
10	1.76bc	5.9cd	15.0abc	17.7a	13.9a	6.5a	2.7bc		
20	1.89abc	6.6bcd	15.6abc	19.5a	14.8a	6.2a	2.9ab		
40	2.15ab	5.6cd	19.6ab	20.6a	15.0a	7.1a	3.4ab		
80	2.21a	5.2cd	15.4abc	18.7a	14.4a	7.7a	3.1ab		
160	1.98abc	3.7d	19.8a	19.0a	14.4a	7.8a	3.7a		

For each response, means sharing the same letter did not differ significantly using Duncan's multiple range test at  $\alpha = 0.05$ .

**Table 2**Mean lemongrass essential oil yield, and the concentrations and yield of caryophyllene oxide (caryo-oxide), neral, geranial and *t*-caryophyllene (*t*-caryo) obtained from the 8 distillation times (DT).

DT (min)	EO yield	Concentration in % of total oil				Yield in mg/500 g biomass			
		Caryo-oxide	Neral	Geranial	t-Caryo	Caryo-oxide	Neral	Geranial	t-Caryo
2.5	0.024d	0.0e	23.6c	24.9c	0.18d	0.0e	28.5e	29.8e	0.2d
5	0.060c	1.0d	31.5ab	31.5abc	0.31d	3.0d	94.4d	94.3d	0.9d
10	0.099b	1.2cd	35.2a	34.6ab	0.36d	6.0c	178.1c	173.8c	1.8d
20	0.163a	1.9bc	31.4ab	30.4abc	0.63cd	15.9b	254.7b	246.9b	5.1cd
40	0.186a	2.5b	36.6a	36.0a	1.12c	23.5a	340.3a	334.4a	10.4bc
80	0.179a	2.5b	27.8bc	28.0bc	2.46b	22.7a	251.7b	253.6b	21.9a
160	0.187a	2.9b	26.7bc	27.4c	1.98b	26.6a	248.3b	254.6b	18.4a
240	0.101b	5.0a	25.5bc	30.4abc	3.22a	24.6a	128.2cd	152.0cd	16.1ab

For each response, means sharing the same letter did not differ significantly using Duncan's multiple range test at  $\alpha = 0.05$ .

with increasing DT, reached maximum at 160 min, and remained unchanged at 240 min DT (Table 3; Fig. 6). Geraniol yield reached maximum at 20 min DT, whereas geranyl acetate yield reached maximum at 160 min DT.

#### 4. Discussion

## 4.1. Effects of DT on essential oil yield

Our findings indicate that for peppermint, lemongrass, and palmarosa essential oils, a DT of 20 min would be sufficient for maximum essential oil yields. In each distillation experiment, we detected no significant increases in oil yields with DTs greater than 20 min. In fact, in lemongrass and palmarosa oils, DTs of 240 min had lower yields than those from 20 to 160 min; over-distillation in these oils led to 25–40% reductions in yield. Since steam distillation in general is not a closed system, it is likely that these reductions are due to evaporative loss of the essential oils. If time and energy are important resources to producers, DT should be 20 min, or between 20 and 40 min (to account for other uncontrollable factors that might play a role).

The range of peppermint oil yield in this study was comparable to previous reports (Zheljazkov et al., 2009). Lemongrass oil yield in this study was lower than in a previous report from the same region (Zheljazkov et al., 2011), due to the fact that in this study we used fresh lemongrass biomass, while in the previous study dried herbage was used (Zheljazkov et al., 2011). Palmarosa oil content was also similar to literature reports from other countries.

## 4.2. Effects of DT on oil composition

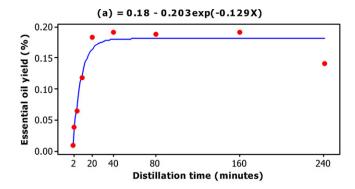
Aside from optimizing yields, DT could be implemented as a tool to engineer essential oils for specific qualities, flavors, or uses. For example, the peppermint essential oil industry in the US consists of producers, dealers, and manufacturers. Manufacturers implement essential oils in various consumer products. To meet a specific targeted aroma, manufacturers request essential oils with a specific profile from dealers. It is the dealers that rectify the various raw essential oils coming from producers by separation and cutting "heads" and "tails" (constituents eluted first or last), to achieve targeted oil characteristics. We demonstrated that varying DT could be

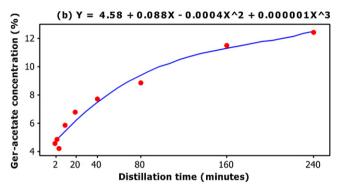
 Table 3

 Mean palmarosa essential oil yield, and the concentrations and yield of geranyl acetate and geraniol obtained from the 9 distillation times (DT).

DT (min)	EO yield %	Concentration in % of total	al oil	Yield in mg/500 biomass	
		Geranyl acetate	Geraniol	Geranyl acetate	Geraniol
1.25	0.009d	4.6e	65.9a	3.1e	44.0d
2.5	0.038cd	4.8e	65.2a	9.1de	121.7cd
5	0.065c	4.2e	63.5a	13.6d	205.6c
10	0.118b	5.8de	71.4a	33.8c	417.8b
20	0.182a	6.8cd	76.8a	61.7b	699.6a
40	0.192a	7.7bc	69.8a	73.6b	666.1a
80	0.188a	8.8b	70.8a	82.5ab	660.7a
160	0.191a	11.5a	66.3a	108.5a	627.3ab
240	0.141b	12.5a	68.1a	86.9ab	477.8ab

For each response, means sharing the same letter did not differ significantly using Duncan's multiple range test at  $\alpha = 0.05$ .





**Fig. 6.** Palmarosa – plot of DT vs. essential oil content and concentration of geranyl acetate (Ger-acetate) together with fitted (a) Asymptotic (concave) for essential oil content nonlinear regression model and (b) third order (cubic) polynomial regression model for Ger-acetate concentration. The fitted models are given as sub-titles.

used as inexpensive method for producing oil with different quality at the producer or dealer level.

Our results indicate that DT can be manipulated to create essential oils with specific properties, within certain limits. Since particular constituents of the three essential oils in this study varied with DT, these constituents can be produced at desired concentrations to achieve various quality goals.

Our data suggest that for peppermint oil, menthofuran and the minor component, eucalyptol, can be modified to meet specific market requirements. Menthofuran, for example, is considered undesirable for some peppermint oil uses, and in that case shorter DTs should be considered. The component t-caryophyllene increased with DT and could be minimized by using shorter DTs, or maximized by using longer DTs. Eucalyptol concentration steadily decreased with DT. Indeed, the yield of eucalyptol remained constant suggesting that eucalyptol completely distills early, but as other components begin to distill, they cause a dilution effect, decreasing overall eucalyptol content in the oil. It is clear that eucalyptol is eluted early; hence, its concentration can be easily manipulated. The initial essential oil product that distills early (within 1.25 min) can be discarded to decrease eucalyptol concentration, or DT can be cut short to obtain oil with increased eucalyptol concentration.

Lemongrass essential oil composition also changed with DT, and therefore, it may be possible to engineer this oil for specific market uses. The major components, neral and geranial were maximized between 10 and 40 min which coincides with DT for optimum yield. The minor components caryophyllene oxide and *t*-caryophyllene both increased with DT. If these components are undesired, using shorter DTs may produce oil with a character dominated by the major components, neral and geranial. Distilling lemongrass at 20 min results in optimization of oil yield, maximization of neral and geranial concentrations, and results in low concentrations of caryophyllene oxide (<2%) and *t*-caryophyllene (<1%).

According to our analysis, shorter distillations of palmarosa oil may result in an essential oil with a more pure geraniol character. If greater concentration of geranyl acetate is desirable for some applications, then palmarosa would need to be distilled for 160 min or longer.

Another important implication for this study is that since oil composition and yield change with DT, it is difficult to compare yields and compositions across published studies; this study can help in that respect. Additionally, different distillation methods result in distinct yields and compositions (Scheffer, 1993) making comparisons even more difficult. For peppermint, various steam distillation studies reporting on yield and/or composition report DTs of 1 h (Zheljazkov et al., 2009, 2010b), 1.5 h (Rohloff et al., 2005), 2 h (Court et al., 1993).

Some authors have reported the yield and composition of palmarosa essential oil without reporting DT (Mallavarapu et al., 1998; Srivastava and Tyagi, 1986), thereby making direct comparison with other studies impossible. Other studies on hydrodistillation and composition of palmarosa oil used DTs of 3 h (Rajeswara Rao et al., 2005; Rajeswara Rao and Rajput, 2011), which is within the range at which we observed the concentration of geranial acetate to increase and essential oil yield to decrease.

#### Acknowledgements

We thank Thomas Horgan, Solomon Green III and Amber Reichley for technical assistance. This research was funded partially by ARS Specific Cooperative Agreement 58-64-2-4-026 with CRIS MIS-172050, specific project "Field Establishment of Medicinal Herbs and Potential for Commercial Production" awarded to V.D.Z. (Jeliazkov).

#### References

Bates, D.M., Watts, D.G., 2007. Nonlinear Regression and its Applications. Wiley, New York.

Cassel, E., Vargas, R.M.F., Joseph, P., 2006. Experiments and modeling of the Cymbopogon winterianus essential oil extraction by steam distillation. Journal of Mexican Chemical Society 50, 126–129.

Court, W.A., Roy, R.C., Pocs, R., 1993. Effect of harvest date on the yield and quality of the essential oil of peppermint. Canadian Journal of Plant Science 73, 815–824. Ganjewala, D., Luthra, R., 2010. Essential oil biosynthesis and regulation in the genus

Cymbopogon. Natural Products Communications 5, 163–172.
Katsiotis, S., Ktistis, G., Zconomou, G.N., 1985. Investigation of the simultaneous influence of three different parameters on the essential oil yield and quality of *Mentha piperita*. Pharmaceutica Acta Helvetiae 60, 228–231.

Koul, V., Gandotra, B., Koul, S., Ghosh, S., Tikoo, C., Gupta, A., 2004. Steam distillation of lemon grass (*Cymbopogon* spp.). Indian Journal of Chemical Technology 11, 135–139.

Kumar, R., Srivastava, M., Dubey, N.K., 2007. Evaluation of Cymbopogon martinii oil extract for control of postharvest insect deterioration in cereals and legumes. Journal of Food Protection 70, 172–178.

Kumaran, A.M., D'Souza, P., Agarwal, A., Bokkolla, R.M., Balasubramaniam, M., 2003. Geraniol, the putative anthelmintic principle of Cymbopogon martinii. Phytotherapy Research 17, 957.

Lawrence, B.M., 2007. Mint: The Genus Mentha. CRC Press, Boca Raton, FL, p. 556. Magee, J.A., Herd, A.C., 1999. Internal standard calculations in chromatography. Journal of Chemical Education 76, 252.

Mallavarapu, G.R., Rao, B., Kaul, P., Ramesh, S., Bhattacharya, A., 1998. Volatile constituents of the essential oils of the seeds and the herb of palmarosa (*Cymbopogon martinii* (Roxb.) Wats. var. motia Burk.). Flavour and Fragrance Journal 13, 167–169.

Montgomery, D.C., 2009. Design and Analysis of Experiments, 7th ed. Wiley, New York.

Rajeswara Rao, B.R., Kaul, P., Syamasundar, K., Ramesh, S., 2005. Chemical profiles of primary and secondary essential oils of palmarosa ((Roxb.) Wats. var. Burk.). Industrial Crops and Products 21, 121–127.

Rajeswara Rao, B.R., Rajput, D.K., 2011. Response of palmarosa [Cymbopogon martinii (Roxb.) Wats. var. motia Burk.] to foliar application of magnesium and micronutrients. Industrial Crops and Products 33, 277–281.

Rohloff, J., Dragland, S., Mordal, R., Iversen, T.H., 2005. Effect of harvest time and drying method on biomass production essential oil yield, and quality of peppermint (*Mentha* × *piperita* L.). Journal of Agricultural and Food Chemistry 53, 4143–4148.

SAS Institute Inc., 2008. SAS/STAT® 9.2 User's Guide. SAS Institute Inc., Cary, NC.

- Scheffer, J., 1993. The isolation of essential oils factors influencing the oil composition. Acta Horticulturae 334, 2–8.
- Srivastava, H., Tyagi, B., 1986. Effects of seed irradiation on yield and quality of essential oil in palmarosa (*Cymbopogon martinii* Stapf.). Euphytica 35, 369–380.
- Weiss, E.A., 1997. Lemongrass. In: Weiss, E.A. (Ed.), In Essential Oil Crops. Cambridge University Press, Cambridge, pp. 86–103.
- Zheljazkov, V.D., Cerven, V., Cantrell, C.L., Ebelhar, W.M., Horgan, T., 2009. Effect of nitrogen, location, and harvesting stage on peppermint productivity oil content, and oil composition. HortScience 44, 1267–1270.
- Zheljazkov, V.D., Cantrell, C.L., Astatkie, T., Hristov, A., 2010a. Yield, content, and composition of peppermint and spearmints as a function of harvesting time and drying. Journal of Agricultural and Food Chemistry, 11400–11407.
- Zheljazkov, V.D., Cantrell, C.L., Astatkie, T., Ebelhar, M.W., 2010b. Peppermint productivity and oil composition as a function of nitrogen growth stage, and harvest time. Agronomy Journal 102, 124–128.
- Zheljazkov, V.D., Cantrell, C.L., Astatkie, T., Cannon, J.B., 2011. Lemongrass productivity, oil content, and composition as a function of nitrogen sulfur, and harvest time. Agronomy Journal 103, 805–881.